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Title: **APT COOLANT PURIFICATION AND PLATEOUT STUDIES**

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APT COOLANT PURIFICATION AND PLATEOUT STUDIES

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ABSTRACT

As part of the Accelerator Production of Tritium (APT) project, experiments were performed at the Los Alamos Neutron Science Center (LANSCE) to provide design data and validate design methods for APT primary coolant purification systems. This paper describes the test facility, preliminary results, and plans for future work. Gamma analyses of water samples show that ion exchange is more efficient than microfiltration for removal of the key spallation product Be-7, which is consistent with previous studies.

INTRODUCTION

The APT plant includes primary coolant purification systems for the target, blanket, window, and shield heat removal systems. For these systems, a fraction of the primary coolant flow is continuously processed through a purification system for removal of radioactivity and control of water quality parameters, including resistivity, dissolved solids, dissolved oxygen, and pH. The following design issues have been identified: (1) Spallation reactions occurring directly in the cooling water produce relatively high levels of 53.3-d Be-7.[a] Deposition of Be-7 has been a concern for operations and maintenance of spallation target cooling systems with Be-7 inventories that are 1000 times or more lower than those expected for the APT plant.[1] (2) Enhanced corrosion rates are expected to occur in regions exposed to high levels of proton flux, which would add radioactivity and particulate matter to the coolant.[2] Radioactivity will also enter the coolant as the result of recoil from wetted metal surfaces following reactions with high-energy protons and secondary particles. (3) Radiolysis may lead to the buildup of longer-lived oxidizing agents (e.g., H_2O_2), which can enhance corrosion of out-of-beam wetted surfaces (e.g., heat exchanger tubes).[3] Activation of these corrosion products is an additional source of particulate matter and coolant activity. (4) There are large uncertainties in the transport and deposition behavior of Be-7, 2.6-y Na-22, and other nuclides that are unique to water-cooled spallation targets. (5) There are large uncertainties in the performance of filters and ion exchange resins for purification of spallation target cooling water. To address these issues, a test was conducted at LANSCE, utilizing its linear accelerator that produces an 800-MeV, 1-mA proton beam.

TEST FACILITY

The test facility consists of an irradiation insert, water cooling system, coolant purification system, plateout probes, a sampling station, and laboratory facilities for analysis of water samples. In addition, ionization chambers and tubes for routing thermoluminescent dosimeters (TLDs) were installed on or near several components of the cooling and purification systems.

Figure 1 shows the helium loop irradiation insert associated with this test, which was one of five inserts irradiated at LANSCE Experimental Area A during the fall of 1998 in support of the APT Engineering Development and Demonstration Program. Figure 2 shows the arrangement of the five inserts. The helium loop insert consists of a lead blanket module and two stainless steel (SS) spallation cylinders, one containing SS tubes and the other containing aluminum tubes. The lead blanket module consists of lead housed within an aluminum plate structure and is designed to obtain irradiation and thermal performance data for the APT lead blanket. The SS and aluminum tubes are designed to obtain data on the chemical

[a] In this paper, half-lives of nuclides are sometimes given before the nuclide abbreviation.

speciation and levels of spallation products in the helium/tritium gas mixture processed by the APT tritium recovery system. A separate helium gas flow system, including gas purification equipment, is connected to these tubes. Within the insert, only SS and aluminum (alloy 6061) surfaces are exposed to the cooling water.

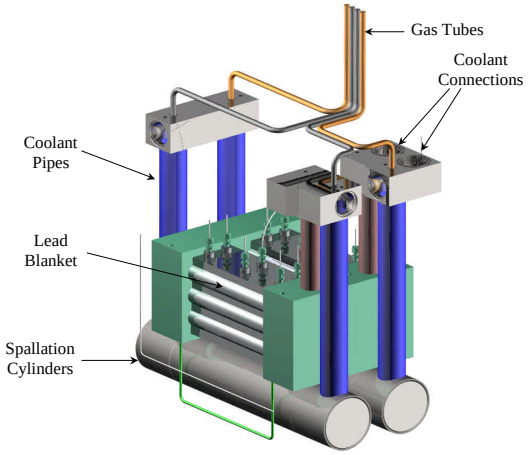

Figure 1. Helium Loop Irradiation Insert

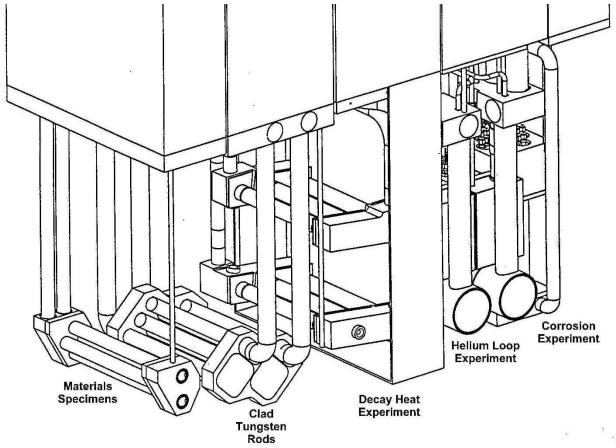

Figure 2. LANSCE Area A Irradiation Inserts, Fall 1998

Figure 3 is a schematic of the water cooling and bypass flow purification systems for the helium loop insert. Instrumentation (not shown on Fig. 3) includes pressure transducers, thermocouples, flowmeters, and level indicators. All valves and pumps are operated remotely, and data acquisition is performed using a LabView® software interface.

Wetted surfaces in the cooling and purification systems consist primarily of 304L and 316L SS, with the exception of materials used for valve seats, seals, and weld filler. Piping for the cooling water system and plateout probe assembly is 1½-inch Schedule 40. Welds were used to connect the components, piping, and valves except at the inlet and outlet of the plateout probe assembly where Swagelok® fittings were used so the entire assembly could be removed with remote handling equipment. The total coolant volume in the system is approximately 70 gallons, and the total system flow rate is about 50 gpm. During most of the test, coolant temperatures were maintained near 60°C by controlling the flow rate on the secondary side of the heat exchanger. The temperature rise through the irradiation insert ranged from 3°C to 5°C at full beam power. System pressures ranged from about 30 psig at pump suction to about 140 psig at pump discharge. Figure 4 shows the water cooling system before final installation and connection to the helium loop insert. The helium loop insert is located on the beam line, about 18 feet below the water cooling system. Steel and concrete shielding keep proton and neutron radiation fields near the water cooling system at very low levels.

The coolant purification system is designed to process approximately 0.5 to 2.5 gpm (1% to 5% of the total flow) through sintered metal filters and ion exchange resin beds. The system accepts flow from the high pressure side of the pump and discharges flow on the low pressure side of the irradiation insert. The system consists of the following components: 2-µm filter, 0.2-µm filter, cation exchange resin tank, mixed bed ion exchange resin tank, and post 2-µm filter. The cation resin is Amberlite® IR-120 in H^+ form, which has proven to be effective for removal of Be-7.[4] The mixed-bed resin is Amberlite® MB-150 in OH^- form. The arrangement of motor-operated valves allows for flow through several different purification system configurations. A flow-control valve is used to adjust the flow rate as required. The components are connected using 0.5-inch diameter SS tubing and Swagelok® fittings. The purification system is located adjacent to the water cooling system and is surrounded by 2½-inch-thick steel shielding. Figure 5 shows the purification system before placement of the shielding.

The water sampling station consists of a steel sink and plexiglas enclosure with water drain and air exhaust systems. Bulk water samples are taken from the surge tank, and purified water samples are taken from the return line of the purification system. Prior to entering the sampling station, sample water is cooled to near room temperature (~25ºC) using a coiled line in a water tank. The sampling station also has a separate line with flow-through cells for resistivity and pH measurements. These cells are connected to a Martek Instruments Mark-22 resistivity/pH analyzer with local readout. A limited number of dissolved oxygen measurements were performed at the sampling station using a custom-designed flow-through cell for use with colormetric ampoules.

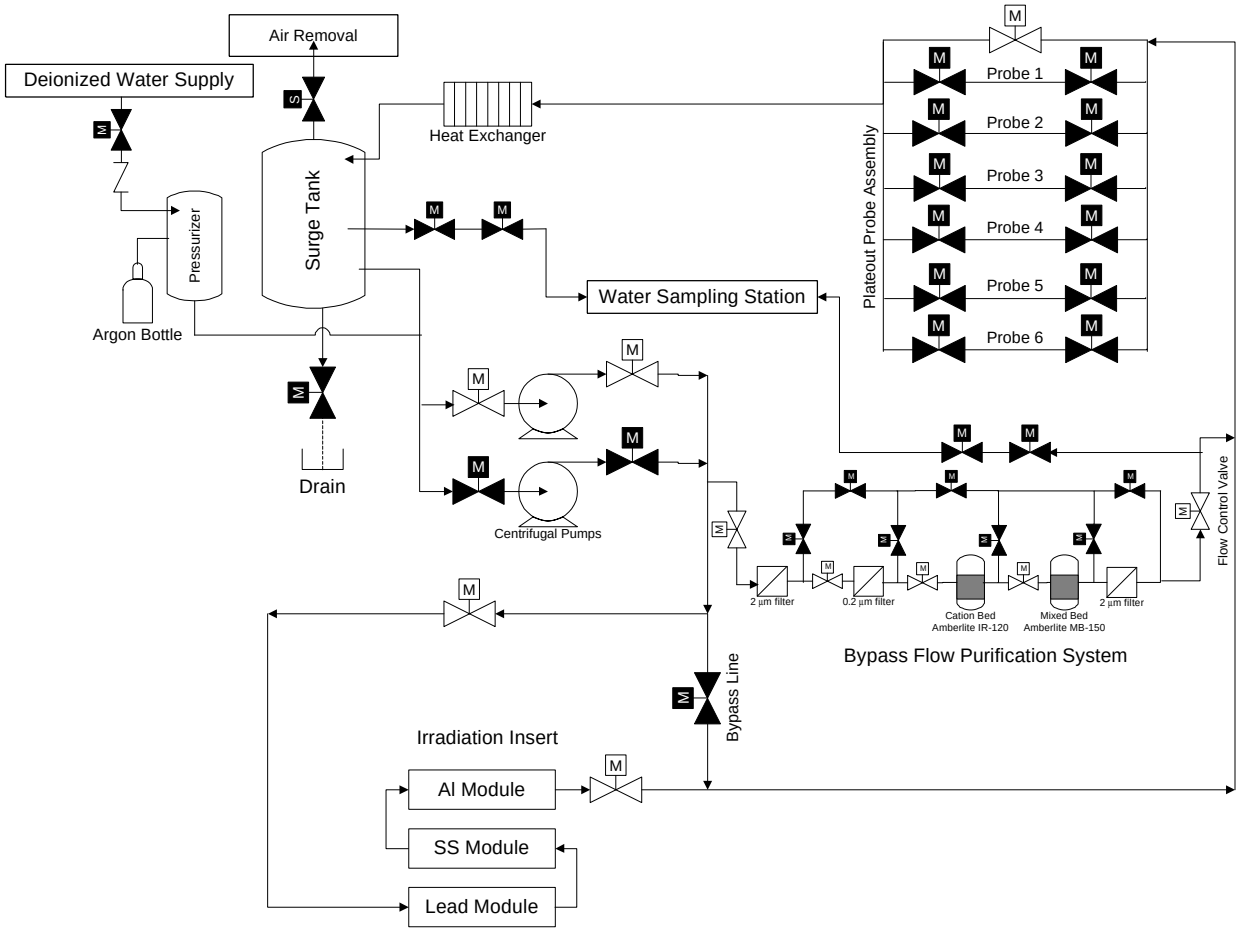

Figure 3. Schematic of Helium Loop Insert Water Cooling and Purification Systems

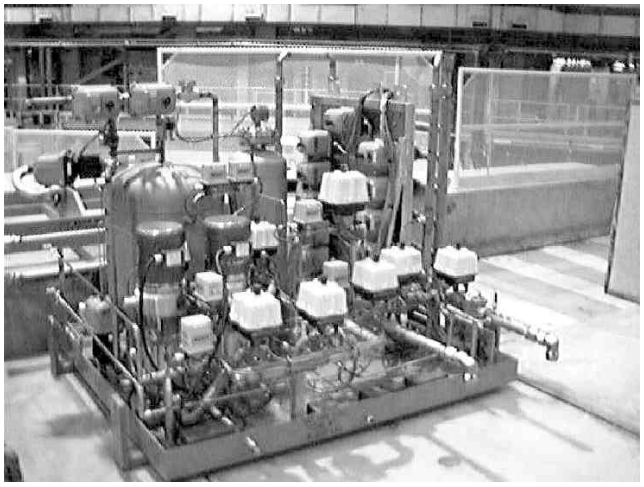

Figure 4. Helium Loop Insert Water Cooling System

Figure 6 shows the plateout probe assembly. The sections of pipe between the isolation valves are approximately 2 feet long. The probes were designed to take radionuclide deposition data under well-characterized conditions, including running the purification system under different configurations to vary the particle size distribution. After a deposition experiment was completed, the water system was drained and the probes were blown dry with argon gas and isolated. Prior to irradiation, the wetted surfaces of probes 5 and 6 were electropolished, and probe 6 was also passivated in nitric acid. The remaining probes were untreated. Probes 4, 5, and 6 were run simultaneously to provide data on radionuclide deposition as a function of surface treatment. These probes were run with the purification system configured for flow through all components. Probe 1 was run with the purification system bypassed; probe 2 was run with flow through all purification system components except the 0.2-µm filter; and probe 3 was run with flow through only the three filters (i.e., both resin tanks were bypassed). As of this writing, the plateout probe assembly has been removed from the water cooling system and replaced with a flexible jumper. The individual pipe sections and valves have been cut from the assembly and are undergoing post-irradiation

examination (PIE). During the PIE, the deposited activity will be leached off the wetted surfaces in order to estimate surface concentrations. These data will be used to develop correlations between coolant and surface concentrations for Be-7 and other key radionuclides.

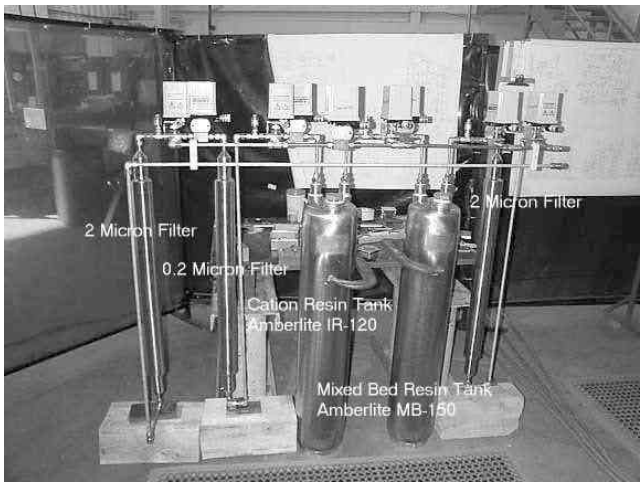

Figure 5. Helium Loop Insert Water Purification System

Figure 6. Plateout Probe Assembly

Ionization chambers were mounted on or near several components in the water cooling and purification systems. The ionization chambers are housed within a polyvinyl chloride (PVC) enclosure, with polypropylene tubes connected to each end of the enclosure. One tube houses the high-voltage cable, and the other tube houses the signal cable. Figure 7 shows the ionization chamber mounted on the heat exchanger.

TLD tubes were also mounted on or near several components and consist simply of polypropylene tubes with Swagelok® caps on the ends. The closed ends are mounted at the desired locations. The tubes are routed through a penetration in the concrete shielding, with the open ends terminating outside the shielding. Dose measurements are performed by attaching a TLD to the end of a flexible cable, pushing the cable through the tube to the capped termination point, and pulling the TLD back through the tube after a prescribed time period has elapsed. Figure 8 shows a TLD tube mounted on the valve of the plateout probe bypass line.

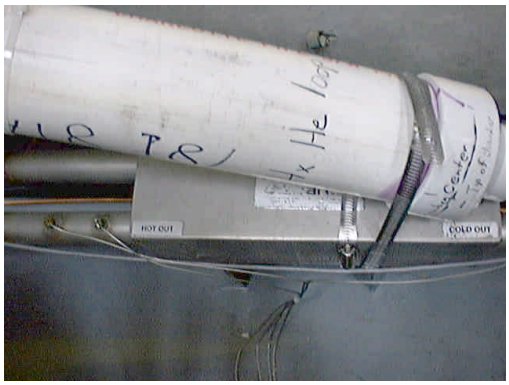

Figure 7. Ionization Chamber Mounted on Heat Exchanger

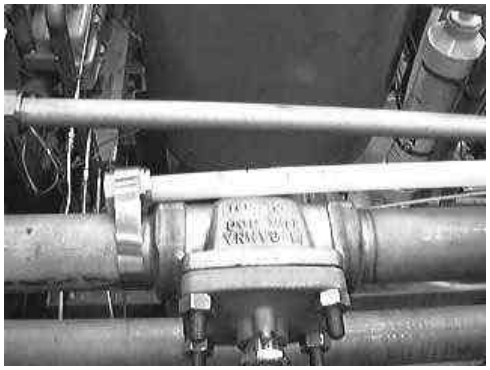

Figure 8. TLD Tube Mounted on Valve of Plateout Probe Bypass Line

In terms of irradiation and flow parameters, the test facility is fairly representative of the APT target primary cooling and purification systems. The test facility is approximately two orders of magnitude smaller in scale than the APT target, but the ratio of beam power to coolant volume for the test facility is within a factor of two of the APT target. The Reynolds numbers for the test facility and APT target are $\sim 2 \times 10^5$ and $\sim 8 \times 10^7$, respectively, but the flow regime is highly turbulent in either case. The transit time for water around the cooling loop is ~1.4 m for the test facility and ~0.8 m for the APT target. The proton fluxes for the two facilities are approximately the same, but the neutron and gamma fluxes for the test facility are an order of magnitude or more below those expected for the APT

target. In addition to SS and Al, a significant portion of the APT target wetted surface area is composed of Inconel 718, which is not present in the test facility. These differences will be taken into account before the test data are used to finalize design of the APT primary cooling and purification systems. For this reason and because analyses are not complete, all data reported in this paper are classified as preliminary.

PRELIMINARY RESULTS

Gamma analyses of water samples clearly showed that the coolant activity was dominated by Be-7. Additional nuclides detected in the water included Na-22, Sc-46, V-48, Mn-52, Mn-54, Co-56, Co-57, Co-58, and Co-60. A few samples were analyzed within several hours after being taken. These samples showed significant levels of 15-h Na-24 and a significant peak at 0.511 MeV from the decay of positron emitters. At the time these samples were counted, the dominant source of positron activity was 20.3-m C-11.

Results are given in Table 1 for Be-7, Na-22, and Co-56 for cases with zero flow through the purification system and with a 5% bypass flow through all components. Operation of the purification system reduces the coolant activity by factors of 60 or more for these nuclides. Later during the experiment, measurements were performed to assess the relative performance of microfiltration and ion exchange. Results are given in Table 2. These results are not directly comparable to those given in Table 1, because several system changes occurred, including repositioning the irradiation insert, increasing cooling water temperature from 48°C to 60°C, and several drain/refill cycles. It is apparent that microfiltration can remove a significant portion of the Be-7, but that ion exchange is a more efficient process, which is consistent with previous laboratory-scale studies.[4] Microfiltration is not very effective for removal of Na-22 or Co-56, indicating that a large portion of the activity associated with these nuclides is in the form of ions or very fine particles.

Table 1. Effect of Coolant Purification on Radionuclide Concentrations

Nuclide	Coolant Activity, μCi/l	
	Purification System Bypassed	Flow Through All Components
Be-7	9600	120
Na-22	4.5	0.025
Co-56	1.4	0.021

Some preliminary measurements of plateout activity were performed by counting swipes taken from the wetted surface of the plateout probe bypass line after the probe assembly was removed and disassembled. These measurements were performed on May 6, 1999, approximately 4½ months after final beam shutdown. Results given in Table 3 show that Be-7 is the dominant contributor to plateout activity, which is consistent with the water sample results.

Table 2. Comparison of Microfiltration and Ion Exchange Purification Performance

Nuclide	Coolant Activity, μCi/l	
	All Components Except 0.2-μm Filter	Filters Only (2 μm + 0.2 μm + 2 μm)
Be-7	34	76
Na-22	0.026	0.28
Co-56	0.016	0.13

Table 3. Gamma-Emitting Radionuclides Detected in Plateout Activity

Nuclide	Half-Life	Surface Concentration μCi/cm^2
Be-7	53.3 d	0.299
Co-56	77.3 d	1.23×10^{-4}
Co-57	271.8 d	2.12×10^{-4}
Co-58	70.88 d	2.43×10^{-4}
Co-60	5.271 y	9.46×10^{-5}
Mn-54	312.2 d	6.62×10^{-4}
Na-22	2.605 y	3.73×10^{-3}
Sb-124	60.2 d	4.79×10^{-4}
Sc-46	83.81 d	8.68×10^{-6}
V-48	15.98 d	8.54×10^{-5}
Y-88	106.6 d	2.60×10^{-4}

An experiment was performed to estimate deposition rates of Be-7 and other key nuclides in the water cooling system. Figure 9 shows results for Be-7. The data points correspond to water samples taken over the time period from December 18, 1998 to January 8, 1999. Final beam shutdown occurred on December 21, 1998 at about 7:15 AM. Flow to the purification system was shut off approximately 10 hours after final beam shutdown. A system drain and refill was performed about one day later, which established baseline conditions for the deposition experiment. Before beam shutdown and drain/refill, the Be-7 coolant activity was 37 μCi/l. Shortly after beam shutdown, the activity dropped to 28 μCi/l. Approximately 10 hours later, the activity was down to 21 μCi/l. This drop

in activity represents the cleanup rate of the purification system, which was configured with a 2.5-gpm flow through all components except the 0.2-µm filter. After about one day of operation with the purification system completely bypassed, the activity increased to 46 µCi/l. This increase is attributed to resuspension of Be-7 from the loop wetted surfaces in the absence of cleanup by the purification system. After the drain/refill process was performed, the activity increased further to 67 µCi/l, which is explained as follows. During the drain/refill process, the bulk of the system water is drained from the tank and loop piping is blown down with argon, but the irradiation insert is bypassed during the blow down and remains filled with the original water. After the system is refilled with clean deionized water, the pump is restarted, but with the irradiation insert bypassed for about 10 minutes to protect turbine flowmeters from bursts of air. During this period, the system flow rate is about 20 gpm higher than with the irradiation insert in line (70 gpm vs. 50 gpm). The higher flow rate increased resuspension of Be-7 at a rate that more than compensated for the loss in inventory from draining. After the drain/refill process was complete, the Be-7 coolant activity decreased at a much faster rate than could be attributed to radioactive decay alone, indicating that deposition was the dominant removal mechanism. Analysis of the data showed the deposition removal constant was about a factor of five higher than the radioactive decay constant. Using this derived deposition rate constant of 2.8×10^{-3}/hr, the fraction of Be-7 deposited per single pass through the loop is 6.5×10^{-5}. Additional analysis is needed before these results can be extrapolated to APT plant conditions. In particular, the effects of differences in Reynolds number, mass transfer coefficients, and loop wetted surface area to coolant volume ratios need to be properly accounted for. Sorption parameters (to be derived from analysis of the plateout probes) are also needed to make meaningful extrapolations.

Figure 10 shows dose rate measurements taken over a 24-hour period using ionization chambers mounted on the heat exchanger and bottom of the surge tank. Also shown is the beam current during this time period. A 4-inch-thick steel plate was placed between the surge tank and heat exchanger to minimize measurement interference. During periods of full-beam current, dose rates from the heat exchanger and surge tank are approximately 50 and 250 rads/hr, respectively, and are dominated by decay of shorter-lived nuclides, including 70.6-s O-14, 122.2-s O-15, and 7.1-s N-16. After beam shutdown, the fairly rapid drop in dose rate is caused primarily by the decay of 20.3-m C-11. During the beam shutdown period, the water system was drained, blown down with argon gas, and refilled with fresh deionized water. With the circulating activity largely removed, the dose rate from plateout activity on the small-volume, high-surface-area heat exchanger is higher than that from activity deposited on the bottom of the surge tank. After beam restart, the dose rates quickly recover to the original levels.

An unexpected result was the relatively rapid buildup of hydraulic resistance across the 0.2-µm filter. Figure 11 shows the associated decay in purification system flow rate over a 24-hour period near the end of irradiation, with the purification system configured with flow through the filters only. This relatively high loading of particulates is probably caused by beam-enhanced corrosion[b] and is confirmed by inductively coupled plasma–atomic emission spectroscopy (ICP-AES) analyses of filtered and unfiltered water samples. For the case with the beam at full power and the purification system bypassed, ICP-AES analyses of bulk system water indicated 2.5 ppm of Al and 2.2 ppm of Fe. The corresponding resistivity was relatively low at 0.04 megohm-cm. This water sample was then split into ten equal parts and filtered through membranes with pore sizes ranging from 0.1 to 10 µm. ICP-AES analyses of acidified membranes showed significant levels of Al and Mg (produced from spallation of Al), but very little Fe. These data indicate the particulate matter may be composed largely of oxides of Al and Mg. Additional analyses are needed to derive the particle size distribution and overall corrosion rate of Al in the system, and to determine if this source of particulates was sufficient to plug the 0.2-µm filter. With the purification system configured for flow through all components, both Al and Fe in bulk system water were below the ICP-AES minimum detection limit of 0.2 ppm; the bulk water resistivity was relatively high at 8.3 megohm-cm; and the resistivity of water taken directly from the return line of the purification system was very high at 13.9 megohm-cm.

Measurements of pH were typically in the range of 4.5 to 6.0, with the lower values corresponding to cases with the purification system bypassed. As discussed previously, a limited number of dissolved oxygen measurements were performed using colormetric ampoules and a custom-designed flow-through cell. Dissolved oxygen levels were in the range of 0.3 to 1.0 ppm. No active measures were employed to control oxygen or other dissolved gases.

FUTURE WORK

Future activities related to these experiments include completion of the PIEs of the plateout probes, additional data analysis, and development of correlations for transport and deposition of Be-7, Na-22, and other key nuclides. The

[b] Beam-enhanced corrosion includes the effects of proton, neutron, and gamma irradiation and radiolysis products.

primary goal is to reduce uncertainties associated with predicting the distribution of radioactivity among the APT circulating primary coolants, wetted surfaces of piping, valves, pumps, heat exchangers, and other components of the cooling systems, and purification system components.

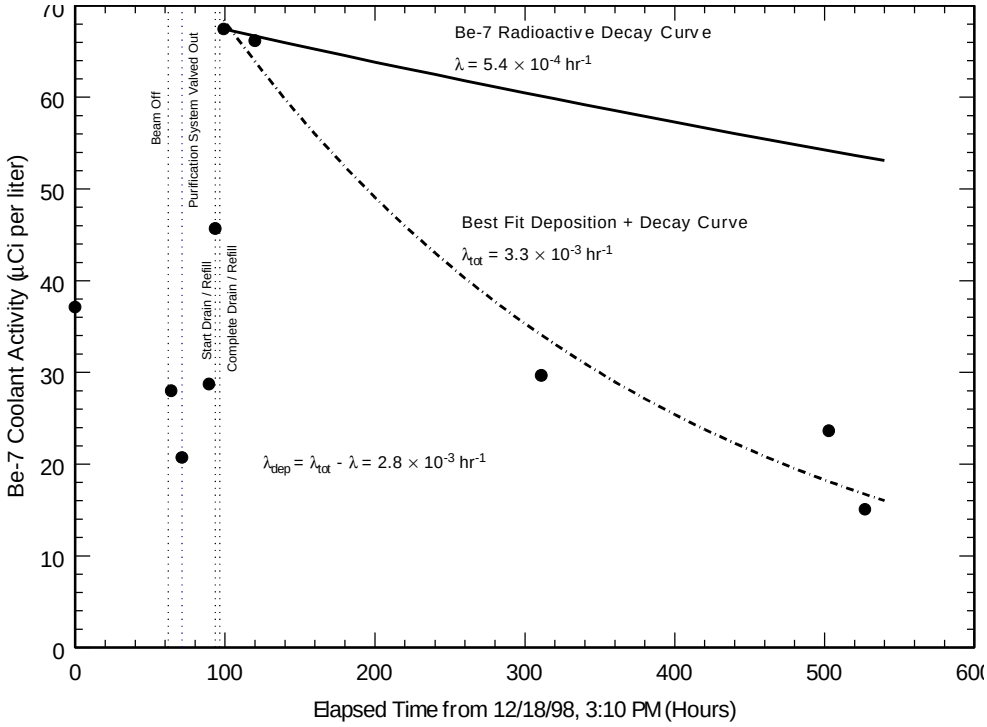

Figure 9. Deposition Behavior of Be-7

Efforts are being made to design the APT primary cooling and purification systems to minimize plateout activity, subject to constraints on practical size limitations of the purification system and associated waste volumes. However, even with an optimized design, it is expected that plateout activity will produce intense radiation fields around APT primary coolant circuits. To that end, a decontamination experiment is being planned for the helium loop insert water cooling system, using commercial processes that have proven to be highly effective for decontamination of light-water reactor primary coolant circuits.[5,6] These processes typically utilize proprietary mixtures of organic acids, chelating agents, and dilute chemicals to control pH. The net result of the process is that the bulk of the plateout activity is transferred to ion exchange resins, with negligible impact on the integrity of the loop wetted surfaces. A high degree of remote maintenance would be required for the primary coolant systems of the APT plant and other facilities that utilize high-power, water-cooled spallation targets unless an effective process is demonstrated for removal of Be-7 and other nuclides that are unique to these systems.

ACKNOWLEDGEMENTS

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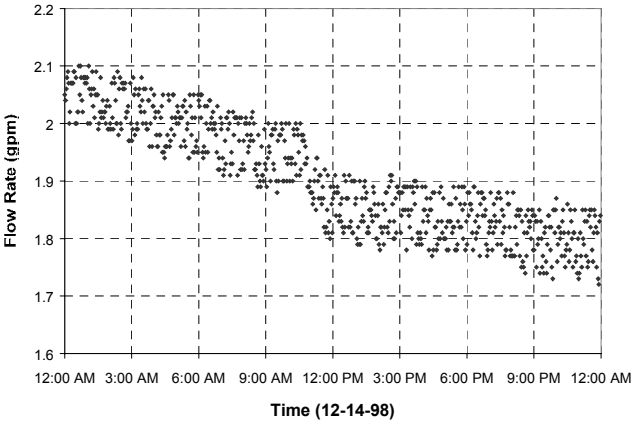

Figure 11. Effect of Particulate Buildup on Purification System Flow Rate

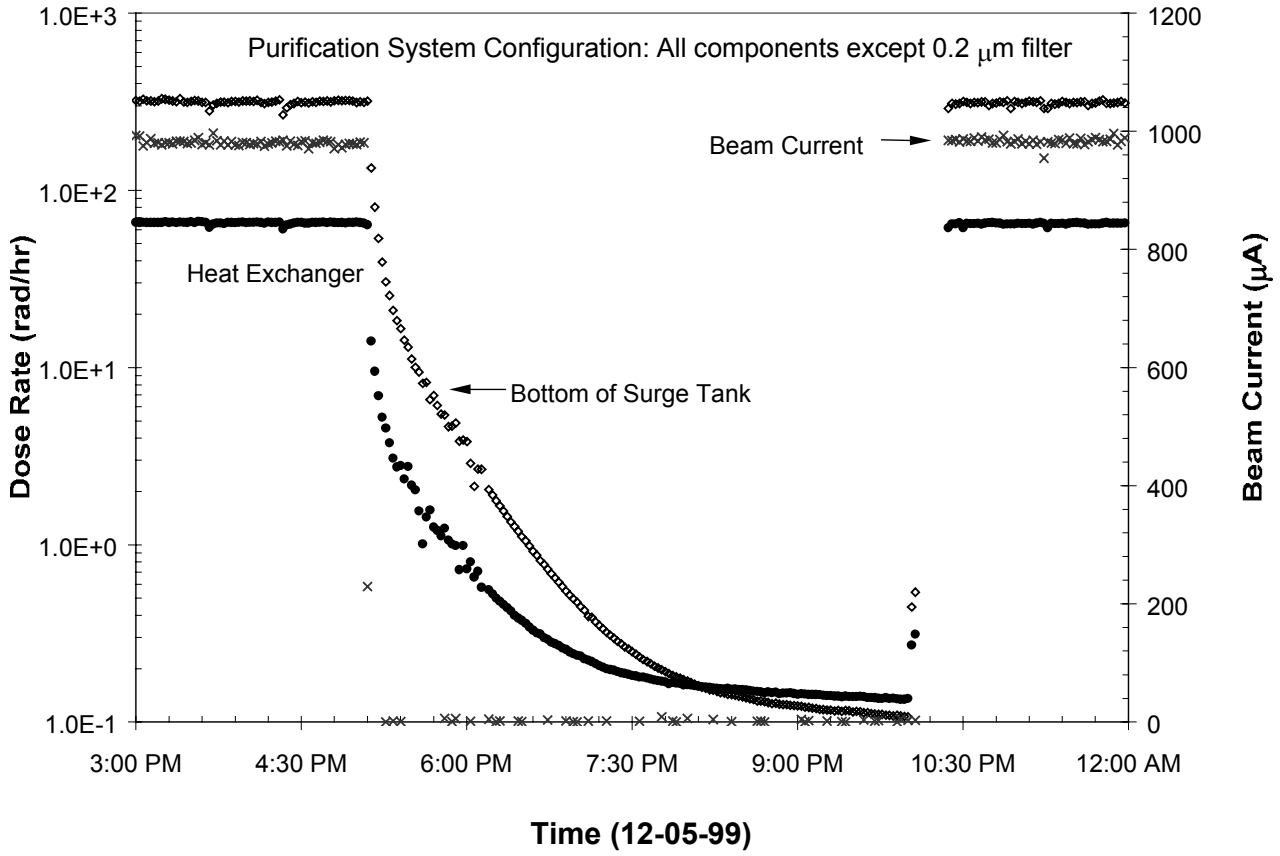

Figure 10. Dose Rate Measurements from Heat Exchanger and Bottom of Surge Tank